Interplay between local electron correlation and Janh-Teller electron-phonon interaction

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The infinite-U Anderson model coupled to a Jahn-Teller phonon is studied using the slave boson method on the basis of the large degeneracy expansion ($1/N$) scheme. The model Hamiltonian acts on the orbital degrees of freedom. The main focus is on the interplay between strong local electron correlation and weak Jahn-Teller electron-phonon interaction. The Kondo temperature is found to decrease by Jahn-Teller interaction. The influence of the Jahn-Teller interaction on dynamical correlation functions is very significant in sharp contrast with the case of the Holstein-type phonon which couples to charge degrees of freedom.

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The orbital degrees of freedom of electrons in a solid play a prominent role in modern material science.[1] In particular, it turns out that electric current can be controlled by manipulating the orbital states of electrons. Spurred by this possibility, a new research area called *orbitronics* is emerging.[2] The orbital physics is also relevant to the quantum transport through orbitally active atoms (or defects) in nanostructures.[3,4]

The dynamics of the orbital degrees of freedom is determined by the Coulomb repulsion between orbitally active electrons and the coupling to the vibrations of ions surrounding the electrons. A well-known example where the orbital physics manifests itself is the transition-metal oxides.[1] Electrons of these materials occupy d orbitals, and the Coulomb repulsion between these electrons is *very strong*. At the same time the electrons in d orbitals are coupled to the vibrations of surrounding oxygen atoms via the so-called Jahn-Teller (JT) coupling.[5] The JT coupling stems from the anisotropy of d orbital wave functions. The *orbital pseudospin* operator to which JT phonon couples is defined by

$$T_\alpha = \sum_\sigma \sum_{a,b} c^\dagger_{\sigma a} \tau^\alpha_{ab} c_{\sigma b}, \tag{1}$$

where σ is a real spin index and a, b are orbital indices. For the case of e_g orbitals of $3d$ electrons, the orbital indices a, b designate two orbitals $d_{x^2-y^2}$ and $d_{3z^2-r^2}$, and the matrices τ^α are the 2×2 Pauli matrices.

The progress of theoretical understanding of orbital physics has been slow in spite of its prime importance. The reason is that the physical phenomena of orbital physics are usually associated with *strong* electron correlation and *simultaneously* with strong coupling to phonons of various types. This makes theoretical analysis exceedingly difficult. Despite these difficulties, the phase diagram of manganese oxides could be qualitatively understood by the mean-field approximation study on the generalized Hubbard model,[6] but the understanding of *dynamical* properties in low energy strong coupling regime is beyond the reach of such mean field approximation. In passing we mention that for *weakly* interacting electron systems there exists a very successful Migdal-Eliashberg theory.[7]

To understand orbital physics in the presence of strong correlation we need the approximation schemes which preserve the essential features of strong correlations. One of such approximation schemes is the dynamical mean-field theory (DMFT).[8,9] In DMFT a lattice Hamiltonian is mapped (or approximated) to a certain *quantum impurity* Hamiltonian. In this brief report we study an Anderson impurity Hamiltonian interacting with local JT phonons (AJT model) from the perspective of the orbital physics within the DMFT scheme or the transport phenomena in nanostructures.

To simplify our problem further *we will assume that real spins are polarized*, namely, the material under consideration is in a ferromagnetic state. This simplification is not unrealistic, since a large portion of phase diagrams of transition-metal oxides and multiferroics are ferromagnetic. A typical example is the low temperature phase of $La_{0.7}Sr_{0.3}MnO_3$ manganese compound with $T_c \sim 370$ K which shows the colossal magnetoresistance.[10] An additional advantage of studying the ferromagnetic state is that we can focus on the orbital dynamics itself not being mixed with spin dynamics.

The physical properties of the AJT model become clearer when they are compared with those of Anderson-Holstein (AH) model.[11,12,13,14,15,16] The dynamical degrees of freedom of the AH model is the *real spin*. The orbital degrees of freedom is not present in AH model. Most important difference between two models is that the Holstein phonon couples to the *charge* degrees of freedom of impurity electron while JT phonon couples to *orbital* degrees of freedom. It is well known that the nonperturbative Kondo singlet ground state of the Anderson model (in local moment regime) is due to the quantum fluctuations in spin (or orbital pseudospin in the context of AJT model) channel. In this regard one can expect that the influence of Holstein phonon on the Kondo ground state would be *small* unless the electron-phonon coupling is extremely large, and this expectation has been confirmed.[12,13,14,15,16] On the other hand the in-

fluence of JT phonon is expected to be substantial since it directly couples to the channel responsible for Kondo (orbital pseudospin) singlet ground state. The results of our study confirm this expectation.

The AJT Hamiltonian consists of the following three parts:

$$\mathcal{H}_{\rm AJT} = \mathcal{H}_{\rm el} + \mathcal{H}_{\rm ph} + \mathcal{H}_{\rm JT}. \tag{2}$$

$\mathcal{H}_{\rm el}$ and $\mathcal{H}_{\rm ph}$ are the Hamiltonians for the electron and phonon parts, respectively. $\mathcal{H}_{\rm JT}$ is the Hamiltonian describing the interaction between the impurity electron and the JT phonon,

$$\mathcal{H}_{\rm el} = \sum_{k,a}(\epsilon_k - \mu_c)\ c^\dagger_{ka}c_{ka} + \epsilon_f \sum_a f^\dagger_a f_a + U f^\dagger_\uparrow f_\uparrow f^\dagger_\downarrow f_\downarrow + \sum_{ka}\frac{1}{\sqrt{N_{\rm lat}}}(V_k f^\dagger_a c_{ka} + V^*_k c^\dagger_{ka} f_a), \tag{3}$$

$$\mathcal{H}_{\rm ph} = \frac{1}{2}\left(\frac{P^2}{M} + M\Omega^2 Q^2\right), \tag{4}$$

$$\mathcal{H}_{\rm JT} = g_0 Q T_z, \quad T_z = \frac{1}{2}(f^\dagger_\uparrow f_\uparrow - f^\dagger_\downarrow f_\downarrow). \tag{5}$$

The indices $a = \uparrow, \downarrow$ denotes the orbital degrees of freedom as $\uparrow = d_{x^2-y^2}$ and $\downarrow = d_{3z^2-r^2}$, and the real spins are assumed to be perfectly polarized. f_a is the local impurity electron operator,and c_{ka} is the conduction electron operator. V_k is the hybridization matrix element, μ_c is the chemical potential for the conduction electrons, ϵ_f is the energy of the impurity level, and U is the local Coulomb repulsion at the impurity site. T_z is the z component of orbital pseudospin. Q is the local Jahn-Teller phonon coordinate. P is the conjugate momentum of Q satisfying $[Q, P] = i\hbar$, and M is ion mass. The JT phonon is assumed to be an Einstein phonon with frequency Ω. $N_{\rm lat}$ is the number of lattice sites for the conduction electrons. g_0 is the electron-phonon coupling constant which is assumed to be *weak*.

¿From Eq. (5) one can notice that the JT phonon plays a role of the fluctuating magnetic field acting on orbital pseudospin. The coupling to the magnetic field evidently hinders the formation of the Kondo singlet ground state, thus the Kondo temperature at which the singlet begins to form is anticipated to decrease. Our results will show that this anticipation is indeed correct. The electron-phonon interaction of the AH model is $\mathcal{H}_{\rm Hol} = g_0 Q\rho$, where $\rho = \sum_a f^\dagger_a f_a$ is the impurity *charge* density, and the index a is the *real spin* index.

The essential features of the Kondo physics of the Anderson model are kept intact in the limit of infinite local Coulomb repulsion U.[17] In this limit the doubly occupied impurity state is not allowed in the physical Hilbert space. The elimination of doubly occupied state provides a considerable formal simplification in the framework of the slave boson method.[18,19,20] Furthermore it possesses the great advantage that it can describe the nonperturbative Kondo singlet ground state at the mean-field level. We study the model Hamiltonian $\mathcal{H}_{\rm AJT}$ in the limit of infinite Coulomb repulsion, employing Coleman's slave boson method in the scheme of $1/N$ expansion[18,21] ($N = 2$ for the original $\mathcal{H}_{\rm AJT}$). In this limit the impurity electron operator f_a can be expressed as

$$f^\dagger_a = s^\dagger_a b, \tag{6}$$

with a constraint $b^\dagger b + \sum_a s^\dagger_a s_a = 1$. b is the slave boson operator, and s_a is a constrained fermion operator. The above constraint is implemented by a complex Lagrange multiplier $\lambda = i\Omega_0 + \lambda_{\rm sa}$. Next we take the Read-Newns gauge in the scheme of $1/N$ expansion in the form $s_a = z_a e^{i\theta}, b = (\sqrt{N/2})re^{i\theta}$. The conduction electron and JT phonon can be integrated out exactly, and then we are led to the following (imaginary time) effective action:

$$\begin{aligned} S = &\int_0^\beta d\tau \sum_m z^\dagger_a(\partial_\tau + \tilde{\epsilon}_f) z_a \\ &+ \int_0^\beta d\tau d\tau' \sum_a \Sigma_0(\tau - \tau')\, z^\dagger_a(\tau) r(\tau) z_a(\tau') r(\tau') \\ &- \frac{1}{2}\int d\tau d\tau' D_0(\tau - \tau')\, \frac{2}{N} g^2 T_z(\tau) T_z(\tau') \\ &+ \int_0^\beta d\tau \sum_a i\Theta\ z^\dagger_a z_a + N\int_0^\beta d\tau\, i\Theta\,(r^2/2 - q) \\ &+ N\int_0^\beta d\tau\, \lambda_{\rm sa}(r^2/2 - q), \end{aligned} \tag{7}$$

where $\tilde{\epsilon}_f = \epsilon_f + \lambda_{\rm sa}, q = 1/N$, and $\Theta = \dot{\theta} + \Omega_0$. Σ_0 is the self-energy due to the interaction with conduction electrons. The imaginary part of Σ_0 is given by $-i\Delta_0 {\rm sgn}(\epsilon)$, where Δ_0 describes the hybrization amplitude which is usually much larger than the Kondo energy scale, while

$$D_0(\tau - \tau') = T\sum_{i\omega} \frac{e^{-i\omega(\tau - \tau')}}{M(\omega^2 + \Omega^2)} \tag{8}$$

is the bare phonon propagator. The fermion field z_a can be integrated out in $1/N$ expansion approximation, and the effective action of bosonic modes r, Θ can be obtained. The parameters $r_{\rm sa}$ and $\lambda_{\rm sa}$ are determined by the saddle point condition, namely that the first order functional derivative of the effective action of bosonic modes vanishes. The Gaussian fluctuations with respect to this saddle point configuration are described by $\delta r = r - r_{\rm sa}$ and $ir_{\rm sa}\Theta$. These Gaussian fluctuations play important role for impurity susceptibilities.[18]

In analogy with the Kondo spin singlet ground state we look for the orbital pseudospin singlet state, which is characterized by the condition

$$\langle z_m(\tau) z^\dagger_n(\tau')\rangle \propto \delta_{mn}. \tag{9}$$

The electron spectral function in saddle point approximation is given by

$$A(\epsilon) = \frac{1}{\pi}\frac{1}{(\epsilon - \tilde{\epsilon}_f)^2 + \Delta^2}, \quad \Delta = \Delta_0 r^2_{\rm sa}. \tag{10}$$

In saddle point approximation the coherent Kondo peak structure is captured, but the incoherent high energy features are missing. ¿From Eq. (10) we can identify Δ with the (renormalized) Kondo energy scale (thus $r_{\rm sa}^2 \ll 1$) and $\tilde{\epsilon}_f$ with the position of Kondo resonance peak. Δ and $\tilde{\epsilon}_f$ are determined by the following saddle point equations (at $T=0$):

$$\lambda_{\rm sa} = \frac{\Delta_0}{\pi}\ln\left[\frac{D^2}{\Delta^2+(\tilde{\epsilon}_f)^2}\right] - \frac{Cg^2}{N}\Delta_0\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}D_0(i\omega)\frac{\partial\Pi(i\omega)}{\partial\Delta} \tag{11}$$

$$\frac{r_{\rm sa}^2}{2} - q = -\frac{1}{2} + \frac{1}{\pi}\tan^{-1}\left(\frac{\tilde{\epsilon}_f}{\Delta}\right) - \frac{g^2C}{2N}\frac{\partial\Pi(i\omega)}{\partial\tilde{\epsilon}_f}. \tag{12}$$

D is the energy cutoff of the order of bandwidth of conduction electron, and C is a numerical constant of order one. $\Pi(i\omega)$ is the polarization function

$$\Pi(i\omega) = -\frac{\Delta}{\pi|\omega|(|\omega|+2\Delta)}\ln\Big[1+\frac{|\omega|(2\Delta+|\omega|)}{\Delta^2+(\tilde{\epsilon}_f)^2}\Big]. \tag{13}$$

If the electron-phonon coupling g is put to zero, Eq. (11) and Eq. (12) reduce to the mean-field equation for the Kondo singlet ground state obtained by Coleman. [see Eq. 2.34 of Ref. 18] The saddle point equations can be solved numerically, but here we are content with the approximate solution for the special case $q=1/2$ ($N=2$). In this case Eq. (12) implies that $|\tilde{\epsilon}_f| \ll \Delta$. This also implies $\epsilon_f \sim -\lambda_{\rm sa} < 0$. Now Eq. (11) can be approximated to

$$\lambda_{\rm sa} \approx \frac{2\Delta_0}{\pi}\ln\frac{D}{\Delta} - \frac{Cg^2\Delta_0}{2}\left[\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}D_0(i\omega)\frac{\partial\Pi(i\omega)}{\partial\Delta}\right].$$

The effect of the interaction with JT phonon is encapsulated in the second term of the above equation. Using the explicit form of $\Pi(i\omega)$ we obtain (η is a numerical constant of order one)

$$\begin{aligned}\Delta &\sim T_K^{(0)}\exp\left[-\eta\frac{E_L}{\Omega}\right] \quad \text{for} \quad \Omega \gg \Delta,\\ \Delta &\sim T_K^{(0)}\exp\left[-\eta\frac{E_L}{\Omega}\Big(\frac{\Omega}{T_K^{(0)}}\Big)^2\right] \quad \text{for} \quad \Omega \ll \Delta,\end{aligned} \tag{14}$$

where

$$T_K^{(0)} \sim D\exp\left[\frac{\pi}{2}\frac{\epsilon_f}{\Delta_0}\right] \tag{15}$$

is the unrenormalized Kondo temperature of the infinite-U asymmetric Anderson model obtained by Haldane using poor man's scaling.[22] E_L is the lattice relaxation energy defined by

$$E_L = \frac{g^2}{M\Omega^2}. \tag{16}$$

This energy scale is also often called the polaron energy. We find that the JT electron-phonon interaction *decreases* the Kondo temperature, which agrees with the previous expectation. For AH model the Kondo temperature *increases* by the interaction with Holstein phonon. The Kondo temperature of the infinite-U AH model obtained with the slave boson method is[16]

$$T_K \sim T_K^{(0)}\left(1+\pi\frac{E_L T_K^{(0)}}{\Delta_0^2}\right),$$

To refine our discussion let us specify the reasonable relative energy scales: $|\epsilon_f| > \Delta_0 > \Omega > (\Delta \gtrsim E_L)$. In this parameter regime the renormalization of Kondo temperature in AJT model is much larger than that in AH model. Apart from the magnitude of renormalization the dependence on various parameters is also markedly different between two models, reflecting the essentially different renormalization mechanism.

It is interesting to attempt to understand the decrease of Kondo temperature by JT interaction from alternative approach. Let us consider the *antiadiabatic limit* where the phonon frequency is the highest energy scale, which in our situation implies that $\Omega > |\epsilon_f| > \Delta_0 > (\Delta \gtrsim E_L)$. Clearly this limit does not have much physical relevance, but it does provide some mathematical simplifications. In the antiadiabatic limit, the bare phonon propagator becomes local in time $D_0(\tau-\tau') \to \delta(\tau-\tau')/M\Omega^2$. Now the integrated JT electron-phonon interaction also becomes local in time (note that $f, f^\dagger$ are Grassman numbers and are *not* operators, in Lagrangian formulation)

$$-E_L T_z T_z \to +2E_L f_\uparrow^\dagger f_\uparrow f_\downarrow^\dagger f_\downarrow.$$

The right-hand side of the above equation is of the same form and of the same *sign* as the local Coulomb repulsion, so that it *increases* the local Coulomb repulsion. To address the renormalization by JT interaction in this setup we had better consider *finite-U* Anderson model. For simplicity let us take the *symmetric* Anderson model. ¿From the Kondo temperature of the symmetric Anderson model[22] with *renormalized* U one can obtain

$$T_K \sim T_K^{(0)}\left(1-2\frac{E_L}{\Delta_0}\right), \tag{17}$$

which is similar to our result Eq. (14) with the replacement of $\Delta_0 \to \Omega$. Even if the antiadiabatic limit is not physical, this result gives a supporting evidence for the validity of our result.

The impurity susceptibilities can be computed by inserting source fields to the effective action Eq. (7) and by integrating out z_a and all bosonic modes.[18] The orbital susceptibility is defined by

$$\chi_{\rm o}(\tau-\tau') = \langle T_z(\tau)T_z(\tau')\rangle. \tag{18}$$

In the Gaussian approximation one can obtain (C is a numerical constant)

$$\chi_{\rm o}(i\omega) = N\Big[-C\Pi(i\omega) + C^2g^2D_0(i\omega)\Pi^2(i\omega)\Big], \tag{19}$$

where $\Pi(i\omega)$ is given in Eq. (13). Even though the details are not presented here, it should be be noted that the fluctuations of bosonic modes $(\delta r, \Theta)$ do not contribute to the orbital susceptibility. Evidently, Eq. (19) are the first two terms of random phase approxiamtion (RPA)-type expansion, so that for small g^2 we can write

$$\chi_{\mathsf{o}}(i\omega) \sim \frac{N}{[-C\Pi(i\omega)]^{-1} - g^2 D_0(i\omega)}. \tag{20}$$

At high frequency $\omega \gg \Omega$ the renormalization by JT phonon becomes negligible. In the low frequency limit $\omega \to 0$

$$\chi_{\mathsf{o}}(i\omega \to 0) \sim \frac{N}{\pi\Delta - E_L}. \tag{21}$$

The result Eq. (21) clearly demonstrates the JT interaction *strongly enhances* the orbital susceptibility. This again reflects the fact that the JT phonons couples to the orbital pseudospin channel directly. In sharp contrast to this result, the *spin* susceptibility of AH model is *not* renormalized by the interaction with Holstein phonon to the same order of approximation.[16]

The charge susceptibility is defined by

$$\chi_{\mathsf{c}}(\tau - \tau') = \langle \delta n_f(\tau) \delta n_f(\tau') \rangle, \tag{22}$$

where $n_f = \sum_a s_a^\dagger s_a = \sum_a z_a^\dagger z_a$ and $\delta n_f = n_f - \langle n_f \rangle$. The fluctuations of bosonic modes play very important role for the charge susceptibility. In the Gaussian approximation we find that in the limit $\omega \to 0$, for the AJT model,

$$\chi_{\mathsf{c}}(i\omega \to 0) \sim \frac{\pi\Delta}{\Delta_0^2}, \tag{23}$$

Apparently the JT interaction seems to have no effect on the charge susceptibility, but this is misleading since the renormalization by JT interaction is reflected in Δ [Eq. (14)]. Thus Eq. (23) signifies that the JT interaction *suppresses* the charge susceptibility. On the other hand, the charge susceptibility at zero frequency of the AH model is given by

$$\chi_{\mathsf{c}}(i\omega \to 0) \sim \frac{\pi T_K^{(0)}}{\Delta_0^2}\Big(1 + \frac{\pi E_L T_K^{(0)}}{\Delta_0^2}\Big),$$

and we can see that the chare susceptibility is *enhanced* but by a very small amount.[16]

In the framework of DMFT there exists a systematic procedure linking the local impurity susceptibility to the susceptibility of the corresponding lattice system.[8,9] In view of the fact that the orbital excitations can be detected by inelastic x-ray scattering, our result on the orbital susceptibility can have important implications for the inelastic x-ray scattering studies of orbital physics. The renormalization of Kondo temperature can be also experimentally checked, for example, by the isotope effects experiments on the transition-metal oxide systems and multiferroics.

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